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Supplementary information

High-entropy ejecta plumes in Cassiopeia A from neutrino-driven convection

In the format provided by the authors and unedited

Peer Review File

Manuscript Title: High-entropy ejecta plumes in Cassiopeia A from neutrino-driven convection

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Referees' comments:

Referee #1 (Remarks to the Author):

Sato et al. present analysis of archival Chandra X-ray data of supernova remnant Cassiopeia A showing evidence for 48Ti in a shocked high-velocity region of Fe-rich ejecta. They connect this element and its abundance ratio to high-entropy buoyant bubbles that help massive stars explode. Understanding the explosion mechanisms of core collapse supernovae is a key topic in stellar evolution and high energy astrophysics, and thus this work has the potential for wide interest among theorists and observers of supernovae and supernova remnants.

Overall the paper is of good quality. However, in my comments below, organized into two major groupings, I describe how the detection as presented is not robust and that additional details must be provided about the methods used. The measurement is critically dependent on careful treatment of the extraction regions and modeling of the continuum. Performing these measurements consistently over multiple epochs of Chandra observations is especially challenging. Moreover, the authors fail to acknowledge prior work that has already directly mapped Cassiopeia A's bubble-like morphology.

As presented the reader cannot have confidence in this result and its interpretation, and I do not recommend this paper for publication in Nature.

[1] How much confidence do the authors have on fitting the continuum in order to measure the excess emission that they associate with 48Ti? The result is critically dependent on this. Ways to substantiate this analysis include addressing the following questions:

---What does the fit look like over the entire observed X-ray spectrum? Only the region between 4 to 7 keV is shown.

---Did the authors consider the Ca XIX line at 4.822 keV?

---How were the extraction regions selected?

---Were other regions investigated? Do the authors have any interpretation about why this one region is rich in 48Ti emission?

---Does changing region boundaries affect the resulting spectrum? And if so, how? There is the concern that changing extraction regions changes the potential for contaminating ejecta.

---Is the large range of n_et a concern for the authors?

Furthermore, the authors need to provide additional details about how data from multiple epochs were coadded. Adding data from multiple epochs of Chandra observations is not trivial. The authors describe following proper motion of the filaments, but as the space craft roll angle changes between epochs, so does the PSF. Consequently, some angles are more blurred than others. The remnant itself is changing as well (see, e.g., Patnaude & Fesen 2009; or lead author's own paper Sato et al. 2018) which complicates unique and constant fitting parameters.

[2] The first sentence of this paper reads that Ni-bubbles have never been directly observed. Yet, there are many papers that have supported the existence of Ni-bubbles and have already mapped their structure *directly* in Cassiopeia A. Among the relevant papers include Milisavljevic & Fesen (2015) who reported observations of a "bubble-like" interior of Cassiopeia A. In their observations, cavities are easily identifiable in [S III] emission at near-infrared wavelengths. These cavities smoothly connect with the greater ring morphology of shocked ejecta visible at optical (Reed et al. 1995; Lawrence et al. 1995; Alarie et al. 2014) and infrared wavelengths (DeLaney et al. 2010). Indications of this structure had been observed by Isensee et al. (2010). The authors make no reference to these papers and fail to comment on how those results connect with their own.

Sato et al. rely on circumstantial evidence based on nucleosynthesis to claim an association between the candidate 48Ti emission and bubbles created by large-scale mixing. However, the protruding fingers do not resemble bubbles at all. Did the authors consider other high entropy processes / instabilities that can create alpha-rich freezeout in this region? DeLaney et al. (2010) considered interaction between ejecta and CSM structures to be plausible in this region.

A citation to Li, McCray, and Sunyaev (1993) is appropriate, as this was among the first papers to describe the Ni-bubble effect. Blondin, Borkowski, and Reynolds (2001) also did foundational computational work on this subject matter. The authors may consider other recent theoretical investigations of this phenomenon, including those by Orlando et al. (2016) and Gabler et al. (2020).

Referee #2 (Remarks to the Author):

Referee report for MS 2020-07-13207

This paper reports the discovery of Ti and Cr in the high velocity Fe-rich ejecta of Cas A. This is interpreted as evidence of high entropy, relatively neutron poor, alpha-rich freezeout conditions in the Fe bubbles.

This paper presents a timely and significant result. It is suitable for publication after some issues are addressed.

The results are sufficiently original. While there is evidence of alpha-rich freezeout in Cas A and other supernova remnants, the spatial information and independent line of evidence facilitate characterization of the supernova engine in a unique way. This information will be relevant to supernova modeling and nucleosynthesis studies. It should be of significant interest to the community. It could also be of interest to the meteoritics community.

Use of statistics and assessment of observational uncertainties appear to be sound.

The observational data and methodology appear to be sound. Including the supplementary information, the x-ray data and fitting are adequately described. There do appear to be discrepancies in figure captions for figures 2, 4, and 5. The captions seem to misidentify the red and black colors. In figures 2 and 4, the captions states the model is black and data points red. In figure 5, the caption states the model with Ti is black.

I have questions about the theoretical methodology. The nucleosynthetic conclusions rely heavily on the characteristics of alpha-rich freezeout. The yields of the freezeout are very sensitive to the thermodynamic evolution of the material, especially for intermediate mass elements. Based on the description of the methods, the quoted abundance ratios are derived from the confined inner layers of one 1D explosion model. The applicability to penetrative Fe bubbles needs to be well supported. The use of a mass cut when examining very small radius, high entropy material with strong overturn also needs to be better justified.

The modeling of the lighter elements as contamination from nearby QSE ejecta is plausible, but it needs to be checked against thermodynamic trajectories for alpha-rich freezeout that produce more intermediate mass elements.

The conclusions appear to be sound with two caveats. First, the paper claims the detection of 48Ti, 52Cr, and 58Ni. The actual detection is of bulk elements. The identity of the isotopes is inferred from nucleosynthetic models. This should be made very clear, especially for the benefit of those outside the supernova field, i.e. meteoriticists, who may find this of interest. Second, some of the conclusions depend strongly on the yields of alpha-rich freezeout. These are highly dependent on the thermodynamic history, but the comparison is based on one 1D explosion calculation. The robustness of the result should be demonstrated with at least yields from a range of analytic trajectories exploring a somewhat wider parameter space. For example, the density of the material in the 1D explosion is not specified, but high Ye material at relatively low densities can produce large amounts of Ti.

Suggested improvements:

Detection of isotopes versus elements: See remarks in conclusiosn section.

In the last paragraph of page 2, the parameters that determine NSE yields are listed. The lepton fraction is more correct than the electron fraction. While under most conditions these are essentially the same, since neutrino chemistry is discussed in the paper, it would be better to be precise. In addition, the paragraph states that the abundances of alpha-rich freezeout elements are determined by these three parameters. This is incorrect. Those three set the abundances for NSE. Freezeout additionally depends on the rate of change of T and rho and the freezeout timescale.

In the second paragraph of page 3, the chemical evolution of Ti/Fe is mentioned. The discussion seems to imply that the Ti/Fe in the high entropy bubbles might help explain the Ti overproduction problem. As the authors note, this is true only if a significant fraction of the Ti and Fe are produced in the high entropy regions. It would help the case if the mass of Ti and Fe in the bubbles were compared to an estimate of the total yield of the explosion.

Additional exploration of alpha-rich freezeout parameter space: please see comments in modeling and conclusions sections.

References are sufficiently complete.

The abstract and body are clear and provide appropriate context with the inclusion of the supplementary material. Specific points requiring clarification are noted above.

Author Rebuttals to Initial Comments:

Referee #1 (Remarks to the Author):

Sato et al. present analysis of archival Chandra X-ray data of supernova remnant Cassiopeia A showing evidence for 48Ti in a shocked high-velocity region of Fe-rich ejecta. They connect this element and its abundance ratio to high-entropy buoyant bubbles that help massive stars explode. Understanding the explosion mechanisms of core collapse supernovae is a key topic in stellar evolution and high energy astrophysics, and thus this work has the potential for wide interest among theorists and observers of supernovae and supernova remnants.

Response: We thank the referee for recognizing the importance of our results. We are also deeply grateful to the referee for indicating the potential for wide interest in this research.

Overall the paper is of good quality. However, in my comments below, organized into two major groupings, I describe how the detection as presented is not robust and that additional details must be provided about the methods used. The measurement is critically dependent on careful treatment of the extraction regions and modeling of the continuum. Performing these measurements consistently over multiple epochs of Chandra observations is especially challenging. Moreover, the authors fail to acknowledge prior work that has already directly mapped Cassiopeia A's bubble-like morphology.

As presented the reader cannot have confidence in this result and its interpretation, and I do not recommend this paper for publication in Nature.

Response: We considered all these points since they are really important for improving our manuscript. And, we conclude that all the concerns can be addressed using the current data set. Some values in the manuscript have slightly changed from the previous version by our reanalysis, but the changes have not affected our conclusions at all.

[1] How much confidence do the authors have on fitting the continuum in order to measure the excess emission that they associate with 48Ti? The result is critically dependent on this. Ways to substantiate this analysis include addressing the following questions:

Response: We are confident in the modeling and analysis. In the Methods section, we demonstrate the robustness of this measurement from various perspectives. Table 5 in the Methods section summarizes our investigations, clearly demonstrating the robustness of our measurement.

1. **The region selection** does not affect our conclusion significantly. We additionally analyzed a different region (dashed contours in Figure 4) and found that the result does not change even with a region reduced in size by roughly half. For more detail, please see the last paragraph in "Chandra observations and data reduction".

2. **The modeling of the continuum emission** is being accurately done. We analyzed spectra from wider energy ranges, finding that the Ti detection significance remains high and the Ti/Fe mass ratio remains consistent. All the lines around the Ti emissions are properly modeled (please see Fig. 7, Table 5 and "Modeling of X-ray spectrum").

3. **The results from the combined multiple epochs of Chandra observations** are consistent with those from a single epoch observation in 2004. The detailed result is shown in "Modeling of X-ray spectrum" and Table 5. Even when using only the single epoch data, we found a significant detection of the Ti line.

—What does the fit look like over the entire observed X-ray spectrum? Only the region between 4 to 7 keV is shown.

Response: We show the entire X-ray spectrum (data and model) in Figure 7 for fits using three different spectral ranges. There is excellent consistency among the three fits in the key energy band of 4–7 keV and thus the Ti/Fe mass ratio does not change among them. Again please see Table 5 and "Modeling of X-ray spectrum" for more detail.

—Did the authors consider the Ca XIX line at 4.822 keV?

Response: Yes. As shown in Figure 6 (which has a detailed summary of lines near Ti), our thermal model does include this Ca line.

—How were the extraction regions selected?

Response: We defined the region using the Fe-K /Si-K ratio image in the 2004 observation to avoid Si-rich regions (Figure 4). The proper motion of the Fe ejecta has been also considered as shown in "Chandra observations and data reduction". We added new Figures and text to clarify the region selection in this section.

—Were other regions investigated? Do the authors have any interpretation about why this one region is rich in 48Ti emission?

Response: Yes, we have investigated other regions (see Figure 16). For example, we have investigated the northern Fe-rich region, where we found a strong Cr line. This suggests that most of the ejecta here could have been produced at the incomplete Si burning (QSE) layer. The QSE layer could also produce Ti, which complicates an explanation uniquely using the high-entropy process for this region. Non-thermal emission is strong in the western region, which makes it difficult to detect the Ti line.

We cannot confidently conclude that only the southeastern region is rich in Ti emission. In fact, we found Ti emission also at the northern region, but we could not conclude that it has an alpha-rich freezeout origin (because of the strong Cr line). Therefore, we have not included that result in the main text. The additional results are now shown in Methods (please see "X-ray spectra in other Fe-rich regions" and Figure 16).

—Does changing region boundaries affect the resulting spectrum? And if so, how? There is the concern that changing extraction regions changes the potential for contaminating ejecta.

Response: We additionally analyzed the data using a smaller region (see the dashed contour in Figure 4, top). We found that changing the region boundaries does not change the Ti/Fe ratio (see Table 5). In addition, as discussed in "The origin of the lighter elements in the iron-rich ejecta" in Methods, we argue that the Si-rich component itself originates in the incomplete Si burning layer which does not contribute significant Ti emission. From these two points, we conclude that the effect of contamination is not a serious concern in our measurements. Please see "Chandra observations and data reduction" and "The origin of the lighter elements in the iron-rich ejecta" for more detail.

—Is the large range of $n_e t$ a concern for the authors?

Response: The large range of $n_e t$ does not affect the detection of Ti and Cr. Fitting the spectrum with a single ionization time plasma model (NEI model), results in a Ti detection with almost the same significance level (Delta Chi-square 25). This is because, as shown in the new panel (left) of Fig. 6, model components with low ionization timescales (10^{10} cm^{-3} s) do not contribute significant line emission. On the other hand, the profile of the Fe Kα line was not well modeled by the single NEI model; to fit this well requires the large range of ionization states expressed with the vvpshock model. The large range of $n_e t$ at the Fe-rich region is also supported by Hwang & Laming (2003).

Furthermore, the authors need to provide additional details about how data from multiple epochs were coadded. Adding data from multiple epochs of Chandra observations is not trivial. The authors describe following proper motion of the filaments, but as the space craft roll angle changes between epochs, so does the PSF. Consequently, some angles are more blurred than others. The remnant itself is changing as well (see, e.g., Patnaude & Fesen 2009; or lead author's own paper Sato et al. 2018) which complicates unique and constant fitting parameters.

Response: We provide more details in Methods. Thanks to this comment, we are more confident about the handling of the multi-epoch data.

The result of using only one epoch (observed in 2004) is presented in "Modeling of X-ray spectrum". Even with only the 2004 data, we detect the Ti line significantly, and the mass ratios are consistent with the full data set. In addition, we newly analyzed all the data excluding 2004, and this subset also produces results consistent with the full data set. We summarize these results in Table 1 and Table 5. This section demonstrates the robustness of our measurements.

As the referee states, the PSF at the location of the Fe-rich plumes would change from epoch to epoch due to differing roll angles. However, the Encircled Energy Radius does not change at the relevant off-axis angle of 3 arcmin. Please see Figure 4.12-13 in https://cxc.harvard.edu/proposer/POG /html/chap4.html. The 90% encircled energy radius at 6.4 keV is 2 arcsec, which is much smaller than the sizes of the regions we used. Thus we conclude that any epoch-dependent PSF effects are unimportant. See the new paragraph in "Chandra observations and data reduction".

[2] The first sentence of this paper reads that Ni-bubbles have never been directly observed. Yet, there are many papers that have supported the existence of Ni-bubbles and have already mapped their structure *directly* in Cassiopeia A. Among the relevant papers include Milisavljevic & Fesen (2015) who reported observations of a "bubble-like" interior of Cassiopeia A. In their observations, cavities are easily identifiable in [S III] emission at near-infrared wavelengths. These cavities smoothly connect with the greater ring morphology of shocked ejecta visible at optical (Reed et al. 1995; Lawrence et al. 1995; Alarie et al. 2014) and infrared wavelengths (DeLaney et al. 2010). Indications of this structure had been observed by Isensee et al. (2010). The authors make no reference to these papers and fail to comment on how those results connect with their own.

Response: We thank the referee for pointing this out; it provides a really important context for our results. We added additional discussion in the main text and Methods ("Previous studies of the ejecta distribution in Cassiopeia A"). All the suggested references are included in the current version.

The new section will help readers to understand the originality of our study and its connections to the existing body of knowledge on Cassiopeia A.

Sato et al. rely on circumstantial evidence based on nucleosynthesis to claim an association between the candidate 48Ti emission and bubbles created by large-scale mixing. However, the protruding fingers do not resemble bubbles at all. Did the authors consider other high entropy processes / instabilities that can create alpha-rich freezeout in this region? DeLaney et al. (2010) considered interaction between ejecta and CSM structures to be plausible in this region.

Response: As the referee points out, there are a number of effects that can create structures like the Fe-rich ejecta in Cassiopeia A. We summarize these here and also in Methods, "Previous studies of the ejecta distribution in Cassiopeia A".

1. Neutrino-driven convection: we have already discussed this scenario in our manuscript.

2. Standing Accretion Shock Instability: this could be another asymmetry during the explosion. However, the SASI is a large scale instability, and it does not seem reasonable for it to reproduce local protruding structures such as the Fe-rich ejecta in Cas A. Thus, although the SASI is likely not the sole cause of the structure, a combination of the SASI and neutrino-driven convection should be able to reproduce these protruding structures. We newly mention this in the main text.

3. Jet-like explosion: this has already been ruled out by the 44Ti observation (Grefenstette et al. 2014, Nature).

4. CSM interaction: if pure CSM interaction made the Fe-rich structure in Cassiopeia A, the outer edge of that structure should be O-rich. This is much different from what is observed (i.e., no low or intermediate mass species ahead of the Fe-rich structure). Although DeLaney et al. (2010) argued "the Fe jet in the southeast occupies a 'hole' in the Si-group emission and does not represent 'overturning,' as previously thought", such structures are in fact well reproduced by multi-dimensional simulations (e.g., Maeda & Nomoto 2003; Orlando et al. 2020). Thus, we conclude that the formation scenario of the Fe-rich ejecta with the inversion of ejecta layers during the SN explosion is more reasonable than the CSM-interaction origin.

To avoid confusion, we replaced the word "bubble" with "plume" in the current version. Our original motivation for using the word "bubble" was not only for explaining its shape, but also to indicate its buoyant behavior. The protruding structures produced by the seed asymmetries arising from convective flow in the neutrino-heated bubble are not necessarily mushroom-shaped, but show finger-like structures (e.g., Hammer et al. 2010). The detailed shape of high-entropy structures (these are the seed asymmetries, not ejecta asymmetries) depends strongly on the resolution of simulations (see Figure 3 in Radice et al. 2016). Therefore, the high-entropy Fe-rich ejecta should not necessarily have bubble-like or mushroom-like shapes. Also, the propagation of the reverse shock through them can change their shape (e.g., Orlando et al. 2020).

A citation to Li, McCray, and Sunyaev (1993) is appropriate, as this was among the first papers to describe the Ni-bubble effect. Blondin, Borkowski, and Reynolds (2001) also did foundational computational work on this subject matter. The authors may consider other recent theoretical investigations of this phenomenon, including those by Orlando et al. (2016) and Gabler et al. (2020).

Response: We thank the referee for introducing these papers to describe the Ni-bubble effect. We agree with the referee's suggestion, however we think the current citation list in the main text is still suitable for our research. Thus, we decide to discuss these previous studies in Methods, "Previous studies of the ejecta distribution in Cassiopeia A". All the suggested references are included in the current version.

Originally, we had considered citing similar papers (e.g., Arnett et al. 1989, Ono et al. 2013) before the 1st submission. The reason why we did not cite them (including Li, McCray and Sunyaev 1993) in the main text is because these simulations were started by "artificially" seeding Rayleigh–Taylor instabilities. Of course, this series of theoretical approaches in the 1990's to explain observations of SN 1987A helped to understand the Ni-bubble effect in CC SNe. On the other hand, more recent simulations in the 2000-2010 decade gave a deeper physical meaning to the seed asymmetries for reproducing the Ni-bubbles, which had a huge influence on our research. Thus, we cite nine papers that discussed the bubble effect by neutrino-driven convection in the main text (Janka et al. 2016; Burrows et al. 2020; Buras et al. 2006; Wanajo et al. 2018; Burrows et al. 1995; Kifonidis et al. 2003; Hammer et al. 2010; Wongwathanarat et al. 2017; Vance et al. 2020).

Referee 2 (Remarks to the Author):

Referee report for MS 2020-07-13207

This paper reports the discovery of Ti and Cr in the high velocity Fe-rich ejecta of Cas A. This is interpreted as evidence of high entropy, relatively neutron poor, alpha-rich freezeout conditions in the Fe bubbles.

This paper presents a timely and significant result. It is suitable for publication after some issues are addressed.

The results are sufficiently original. While there is evidence of alpha-rich freezeout in Cas A and other supernova remnants, the spatial information and independent line of evidence facilitate characterization of the supernova engine in a unique way. This information will be relevant to supernova modeling and nucleosynthesis studies. It should be of significant interest to the community. It could also be of interest to the meteoritics community.

Response: We are deeply grateful to the referee for indicating the potential for wide interest in this research. In addition, we thank the referee for providing us fruitful comments to revise this paper. In particular, suggestions for investigating the yields with a wider parameter space were very important for our research, making our results potentially more impactful. We have made some changes in the main text based on the parameter study and additional analyses as listed below.

1. When we consider a wider parameter space, the mass fraction (especially Ti/Fe and Cr/Fe) in the Fe-rich ejecta well agrees with proton-rich nucleosynthesis. (Please see new Figure 3 and "Parameter studies of nucleosynthetic outputs in the peak temperature-density plane" in Methods)

2. We now argue that the high-entropy and proton-rich environment is more favorable for accounting for the Mn/Fe mass ratio than the neutron-rich environment. The Mn fraction is effectively produced in the proton-rich environment (see Figure 10 in Wanajo et al. 2018) and also sensitive to the radiation entropy (see Figure 12). We found the Mn/Fe mass ratio agrees with the proton-rich ejecta very well. For more detail, please see "The amount of Manganese in the Fe-rich ejecta" in Methods. We found that the Ni/Fe mass ratio used in the previous discussion is not so sensitive to the proton-rich environment (Figure 15b) and has a large uncertainty, making discussion of the Ye value less compelling in this work. Please see "Another strong evidence of α-rich freeze out" in Methods for more detail.

3. The discussion about the Ti underproduction problem is moved from the main text to Methods. In its place, we discuss the results of the parameter study and the relevance to previous studies (based on the comments by the other referee).

4. The observed values are slightly updated based on additional analysis.

Use of statistics and assessment of observational uncertainties appear to be sound.

The observational data and methodology appear to be sound. Including the supplementary information, the x-ray data and fitting are adequately described. There do appear to be discrepancies in figure captions for figures 2, 4, and 5. The captions seem to misidentify the red and black colors. In figures 2 and 4, the captions states the model is black and data points red. In figure 5, the caption states the model with Ti is black.

Response: We revised the captions correctly.

I have questions about the theoretical methodology. The nucleosynthetic conclusions rely heavily on the characteristics of alpha-rich freezeout. The yields of the freezeout are very sensitive to the thermodynamic evolution of the material, especially for intermediate mass elements. Based on the description of the methods, the quoted abundance ratios are derived from the confined inner layers of one 1D explosion model. The applicability to penetrative Fe bubbles needs to be well supported. The use of a mass cut when examining very small radius, high entropy material with strong overturn also needs to be better justified.

The modeling of the lighter elements as contamination from nearby QSE ejecta is plausible, but it needs to be checked against thermodynamic trajectories for alpha-rich freezeout that produce more intermediate mass elements.

Response: Please see the new Figure 3 and "Parameter studies of nucleosynthetic outputs in the peak temperature-density plane". We did a parameter study like Magkotsios et al. (2010). In Figure 11b, we also show results for different thermodynamic evolution along power-law trajectories. As the referee pointed out, we found the yields of Ti and Cr to vary among the different thermodynamic trajectories. We could not implement some special thermodynamic trajectories realized in multi-dimensional simulations, but we referred to papers (e.g., Wanajo et al. 2018; Vance et al. 2020) that discuss it.

The conclusions appear to be sound with two caveats. First, the paper claims the detection of 48Ti, 52Cr, and 58Ni. The actual detection is of bulk elements. The identity of the isotopes is inferred from nucleosynthetic models. This should be made very clear, especially for the benefit of those outside the supernova field, i.e. meteoriticists, who may find this of interest.

Response: We made a few corrections related to this comment. We show only the bulk elements in the main text. This is because the dominant isotopes are different between neutron-rich and proton-rich ejecta. We found it was difficult to argue for the detection of specific isotopes. On the other hand, in Methods, we show all the isotopes in our models (see "Nucleosynthesis model calculations" and Table 3). this section will help readers who want to know more details about the isotopes.

Second, some of the conclusions depend strongly on the yields of alpha-rich freezeout. These are highly dependent on the thermodynamic history, but the comparison is based on one 1D explosion calculation. The robustness of the result should be demonstrated with at least yields from a range of analytic trajectories exploring a somewhat wider parameter space. For example, the density of the material in the 1D explosion is not specified, but high Ye material at relatively low densities can produce large amounts of Ti.

Response: This was an important comment for us. As we mentioned in previous replies, we did carry out a parameter study. We found that our original 1D SN calculation was limited in expressing the extreme environment in the hot bubbles produced by neutrino heating. We are deeply grateful to the referee for pointing this out.

Suggested improvements:

Detection of isotopes versus elements: See remarks in conclusiosn section.

Response: Revised as mentioned above.

In the last paragraph of page 2, the parameters that determine NSE yields are listed. The lepton fraction is more correct than the electron fraction. While under most conditions these are essentially the same, since neutrino chemistry is discussed in the paper, it would be better to be precise. In addition, the paragraph states that the abundances of alpha-rich freezeout elements are determined by these three parameters. This is incorrect. Those three set the abundances for NSE. Freezeout additionally depends on the rate of change of T and rho and the freezeout timescale.

Response: We revised these sections; please see the text.

In the second paragraph of page 3, the chemical evolution of Ti/Fe is mentioned. The discussion seems to imply that the Ti/Fe in the high entropy bubbles might help explain the Ti overproduction problem. As the authors note, this is true only if a significant fraction of the Ti and Fe are produced in the high entropy regions. It would help the case if the mass of Ti and Fe in the bubbles were compared to an estimate of the total yield of the explosion.

Response: We moved this discussion (Ti overproduction problem) into Methods. As the referee pointed out, we need a significant fraction of the Ti and Fe to be produced in the high entropy regions to explain the Ti overproduction. We now discuss it by referring to a recent multi-dimensional simulation that achieved a quite high Ti/Fe ratio of 10^{-2} (Vance et al, 2020). In our rough calculation, 40% of the Fe-rich ejecta (α-rich freezeout products) must be produced in such an environment. We believe that future multi-dimensional simulations will help make further progress on this question.

Additional exploration of alpha-rich freezeout parameter space: please see comments in modeling and conclusions sections.

Response: We investigated the nucleosynthetic outputs with a wide parameter space (please see "Parameter studies of nucleosynthetic outputs in the peak temperature-density plane" in Methods, Figure 3 and Figure 10).

References are sufficiently complete.

The abstract and body are clear and provide appropriate context with the inclusion of the supplementary material. Specific points requiring clarification are noted above.

Response: We addressed all the comments.

Reviewer Reports on the First Revision:

Referees' comments:

Referee #1 (Remarks to the Author):

The authors have done an outstanding job responding to all referee concerns and suggestions. The thoroughness of the analysis is truly impressive and the paper as presented now is of very high merit. I strongly recommend that this paper be accepted for publication in Nature.

Referee #2 (Remarks to the Author):

The revisions to the manuscript regarding theory and nucleosynthesis satisfy my recommendations and address my concerns. I will leave the responses on the observational questions to my fellow referee, who is more of an expert. If the observational comments are adequately addressed, I believe the manuscript is now suitable for publication.

Patrick A. Young

Author Rebuttals to First Revision:

Referee #1 (Remarks to the Author):

The authors have done an outstanding job responding to all referee concerns and suggestions. The thoroughness of the analysis is truly impressive and the paper as presented now is of very high merit. I strongly recommend that this paper be accepted for publication in Nature.

Response: We are deeply grateful to the referee for his/her careful review and recommending that our paper be accepted for publication in Nature. His/her comments were very important to improve the observational parts in our manuscript.

Referee 2 (Remarks to the Author):

The revisions to the manuscript regarding theory and nucleosynthesis satisfy my recommendations and address my concerns. I will leave the responses on the observational questions to my fellow referee, who is more of an expert. If the observational comments are adequately addressed, I believe the manuscript is now suitable for publication. Patrick A. Young

Response: We are deeply grateful to the referee for recommending that our paper be accepted for publication in Nature. His/her comments were very important to improve the manuscript regarding theory and nucleosynthesis.